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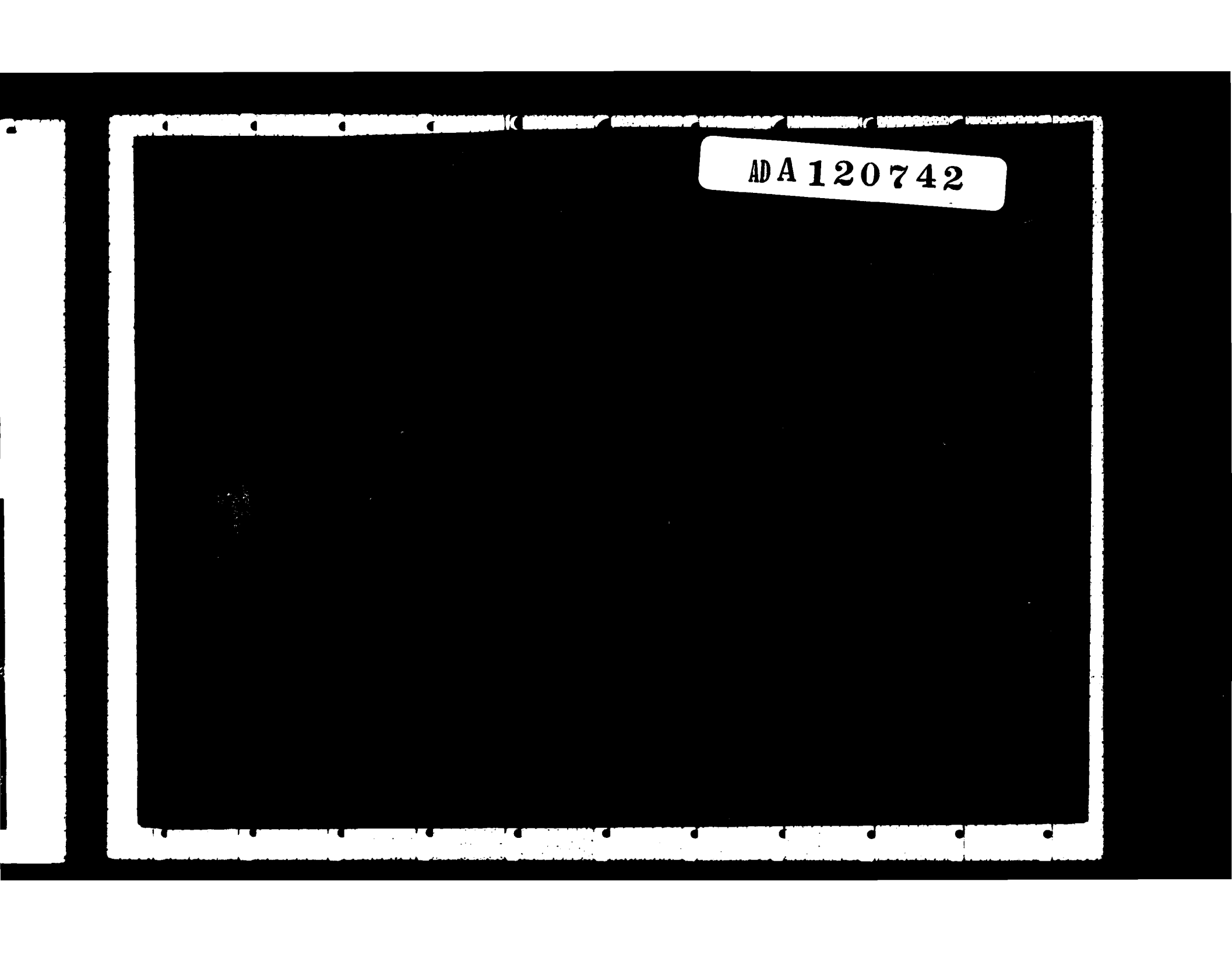

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ENERGY AND MOMENTUM ACCOMMODATION COEFFICIENTS ON PLATINUM AND SILVER

by

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Acoustical measurements of energy and tangential momentum accommodation coefficients indicate stable and reproducible Pt and Ag surfaces are obtained by heating in a vacuum. EAC and TMAC values are given for He and Ne on the heat-cleaned Pt and Ag surfaces and for these gases on Pt and Ag surfaces after exposure to various adsorbent gases. AC values are also given for the various adsorbent gases on themselves on the heat-cleaned Pt and Ag surfaces.

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ABSTRACT

Acoustical measurements of energy and tangential momentum accommodation coefficients indicate stable and reproducible Pt and Ag surfaces are obtained by heating in a vacuum. EAC and TMAC values are given for He and Ne on the heat-cleaned Pt and Ag surfaces and for these gases on Pt and Ag surfaces after exposure to various adsorbent gases. AC values are also given for the various adsorbent gases on themselves on the heat-cleaned Pt and Ag surfaces.

I. INTRODUCTION

When a sound wave is propagated through a gas contained in a cylindrical tube it experiences an absorption and reduction in velocity due to thermal and viscose losses at the tube wall. Approximate analytic equations for the "tube absorption" and "tube velocity" were developed by Kirchhoff[1] assuming a zero particle velocity and sound temperature variation at the tube wall. To arrive at the analytic expression, for these losses, Kirchhoff used a one-term expansion of Bessel functions. In order to explain more recent measurements of sound velocity and absorption in gases at low pressures, it has been necessary to use a numerical solution of the wave equation and to assume a "temperature jump" and "slip velocity" at the tube wall.[2,3] The temperature jump and slip velocity, in turn, are a function of the energy and tangential momentum accommodation coefficients. (See footnote 12 of reference 3.) The sound absorption and velocity, therefore, are a function of the energy and momentum accommodation coefficients, and it is possible to obtain EAC and TMAC values by varying them until the calculated sound velocity and absorption agree with measured values. This method has been used to observe the change in EAC and TMAC for the Ne and He on W as adsorbed O_2 is removed from a tungsten surface by flashing.[4,5] More recently, the method has also been used to compare EAC and TMAC for different W surfaces and for a W surface with different types of molecules adsorbed on the surface.[6,7]

The two parameters, EAC and TMAC, enter into the equations for sound velocity and absorption in a similar way. For this reason, fitting the theoretical expression for sound absorption and velocity to experimental measurements gives the sum of EAC and TMAC more accurately than their individual values.[6] Nevertheless, the acoustical measurements were some of the first to show that TMAC experiences a dramatic decrease similar to that of EAC when adsorbed molecules are removed from a metal surface. (See footnote 2 of reference 6.)

It has proven convenient to display the experimental measurements in terms of the dimensionless parameters C_0/C and $\alpha C_0/\omega$, where α is the absorption coefficient, C_0 is the ideal-gas, free-space sound velocity, and ω is the angular sound frequency. These correspond to the real and imaginary parts of the "reduced" propagation constant. These are a function of the accommodation coeffieients, the ratio of specific heats and the two reduced parameters r' and r_ω; where

$$r' = r\omega/C_0$$

and

$$r_\omega = C_0{}^2\rho_0/\omega\eta.$$

Here r is the tube radius, ρ_0 is the gas density, and η is the gas viscosity.

The sound measurements are generally made at approximately constant frequency as the pressure is varied. EAC and TMAC are obtained, therefore, by plotting the reduced inverse velocity and the reduced absorption as a function of r_ω for a fixed r'. EAC and TMAC are then varied by a computer program to give theoretical curves best fitting the experimental points.

II. EXPERIMENTAL

The experimental system used for the measurements was the same as that used for the measurements on the tungsten tubes.[4-7] The transducers previously used were replaced by capacitance microphones specially made by the Brüel and Kjaer company without the use of organic materials. The sound frequency was approximately 20 kHz, which was slightly higher than that previously used. All of the test gases except CO_2 were passed through a liquid nitrogen trap before entering the measuring system. However, checks indicated this had little effect on the results. The titanium purifier used in previous measurements was removed with negligible effect upon the results.

The Pt and Ag tubes were supplied by the Englehard Industries, Inc. They had an outside diameter of 0.9525 cm and 0.0254 cm wall. The inner surface was

specified as having a 2.5×10^{-5} r.m.s. cm finish and the metal as having 99.95% purity. Electron micrographs of the tube surfaces before and after heating are shown in Fig. 1 A-D. The gases were the same as those previously used on the tungsten tube.

The system was baked out at 150°C for several hours and the pressure was pumped down to the order of 10^{-7} Torr before measurements began. Misch metal getter was evaporated inside the vacuum system before the tubes were heated by induction heating to remove adsorbed gases from the metal surface.

III. DISCUSSION OF RESULTS

A. Measurements with the Pt tube.

Fig. 2 shows the reduced inverse velocity and the reduced absorption data for Ne and He on Pt. The filled stars are the average of three runs taken between July 2 and 9, 1982. The tube was heated in the presence of misch metal getter at 1500 K several times before the measurements on July 2 and also on July 9. Allowing the tube to stand for four days in a vacuum of about 10^{-7} Torr in the presence of misch metal getter produced no observable change in the surface. Furthermore, additional heating of the tube at 1800 K produced no observable additional change in the surface. While it is probably true that the poor vacuum did not allow the obtaining of a clean surface, the surface that was obtained by the heating in a vacuum was stable and reproducible.

Also shown in Fig. 2 are measurements made in He and Ne gas after the Pt tube had been exposed to 10 Torr of various gases. The procedure was to begin with the heat-cleaned surface, then introduce the adsorbing gas (O_2,N_2,CO or CO_2) and measure the sound velocity and absorption in it. This adsorbing gas was then pumped out and the inert gas (He or Ne) introduced into the tube and measured. The tube was then heat-cleaned in a vacuum before introducing the next adsorbent.

As seen in Fig. 2, the exposure of the surface to N_2 produced a negligible

change in the absorption and velocity. The conclusion here is that the N_2 does not stick to the Pt surface. Since the exposure to N_2 produced no change in the surface, it was not heat-cleaned before introducing the next adsorbing gas which was CO.

Table I. records the TMAC and EAC values obtained by fitting theoretical curves to the experimental data. Values obtained from seven runs with the heat-cleaned surface and the one run after exposure to N_2 were averaged. The first two rows in the table give these average values. Measurements after the surface had been exposed to each of the other three gases (O_2,CO,CO_2) gave results differing significantly from those with the heat cleaned surface but differing from each other by less than experimental error. The A.C. values obtained for these runs were averaged and the average values entered in Table I. The ± values in the Table indicate scatter in the data for the surfaces that are grouped together. This scatter is estimated to be within the absolute error of the individual measured values.

For the measurements with the W tube reported earlier, only the sum TMAC + EAC was obtained from the acoustical measurements and the individual values for the two were obtained by comparing the results with EAC values report in the literature.[6,7] This time the measurements are accurate enough to justify reporting both TMAC and EAC. However, it is still true that the sum of the two is determined by the acoustical method with considerably more accuracy than either one individually. The curves shown in Fig. 2. were calculated using the AC values given in Table I.

After exposing the Pt to O_2 it was first flashed at 1100 K and then subsequently at 1400 K. Although the 1100 K flashing temperature lowered TMAC and EAC values considerably (0.60 and 0.12 for He and 0.64 and 0.28 for Ne) the higher flashing temperature was necessary to restore the AC values measured before the O_2 was introduces. In the case of CO and CO_2, the adsorbed molecules were removed (i.e., the original AC values were restored) by flashing at 1100 K.

After the measurements with the Ag tube, the Pt tube was returned to the

system and additional measurements made to determine the effect of a two-week exposure to the atmosphere. These results are also plotted in Fig. 2 and clearly indicate a surface with greater accommodation coefficients than that produced by the exposure to the 10 Torr of the individual gases. The AC values for this surface are also given in Table I. It should be noted that measurements with both the Pt and Ag tubes before they were heat-cleaned for the first time gave AC values greater than any in Table I.

An effort was made to clean the Pt surface by ion bombardment. For this purpose, the Pt tube was suspended in the vacuum system co-axially around the 0.51 cm shaft that carried the sound source. Misch metal getter was deposited on the shaft before it was extended through the Pt tube. A glow discharge was maintained in 10 Torr of A for 20 minutes by a difference of potential of approximately 200 volts between the shaft (which was positive) and the tube (which was negative). The current was varied from 100 to 50 ma to maintain a discharge that filled the tube. This process produced negligible change in AC values measured for the surface.

B. Measurements with the silver tube.

Because of its relatively low melting point (1234 K) and low emissivity it was not possible to raise the flashing temperature of the silver tube high enough to accurately measure it with the optical pyrometer. This temperature was estimated to be 900 ± 150 K. The tube was held at this temperature for a few minutes. The glass wall of the vacuum system was darkened slightly in the heating process, presumably by the deposition of a thin coating of Ag. The procedure for the measurements with the Ag tube was similar to that with the Pt. This time only O_2 produced an observable change in the surface and it was removed by the 900 K flash. Fig. 3 shows the reduced absorption and velocity curves for the silver tube, and Table I gives TMAC and EAC values as determined by fitting the theoretical curves to the experimental points. As with the Pt tube, instead of trying to show different values for each adsorbant, the results for sets of data differing by less than experi-

mental error have been grouped together and averaged. The + or - values in the table indicate the limits of deviation of individual measurements from the average value. If the entry in the table is for a single surface, the ± value is obtained from the estimated accuracy of the velocity and absorption data.

Also given in Table I are the TMAC and EAC values for the various adsorbent gases on themselves on the two metals. Measurements were not possible in CO_2 because the 20 kHz frequency used was above the cut-off frequency for the first non-plane mode in the tube.

IV. COMPARISON WITH OTHER MEASUREMENTS

Very few measurements of TMAC on controlled surfaces have been reported. Lord and Thomas[8] working with vacuum deposited surfaces report values of 0.35 and 0.25 for He and Pt and Ag respectively, values considerably below those reported here.

A number of EAC measurements of Pt surfaces have been made, mostly with thermal conductivity cells. Saxena and Joshi[9] have recently summarized these measurements. As might be expected, values vary greatly (from 0.026 to 0.70) depending upon the nature and history of the surface. Faust,[10] one of Thomas' students at the University of Missouri, made measurements on heat cleaned Pt in the presence of Al getter. His "clean surface" He and Ne EAC values (0.055 and 0.28 respectively) were in relatively good agreement with those reported in Table I (0.07 ± 0.03 and 0.20 ± 0.05). It is not hard to find EAC values for gas covered Pt close to the He and Ne values reported here (0.24 ± 0.08 and 0.47 ± 0.05 respectively). Thomas and Golike[11] measure 0.18 and 0.43 with the temperature jump method.

Most earlier measurements of EAC for CO, N_2 and O_2 on Pt report values 0.77 ± 0.03,[8] which is close to the average value for these three gases given in Table I.

Few measurements have been made of EAC values on a silver surface. Those that have been r[illegible]ed [illegible]vide little basis for comparison with results here reported.

V. CONCLUSIONS

The following conclusions can be drawn from these experiments:

1. N_2 does not stick to the heat-cleaned Pt surface, but CO, CO_2 and O_2 do. These three adsorbing gases have similar effects on the AC values.
2. CO and CO_2 are removed from the Pt surface by heating to 1100 K. O_2 was only partially removed at this temperature. 1400 K removed the remaining O_2 and 1800 K produced no additional change in the Pt surface.
3. For the silver tube, exposure to neither CO nor N_2 changed the AC values. O_2 was removed by heat cleaning at approximately 900 K.
4. To within experimental error, a single TMAC value of 0.75 has been observed for both He and Ne on the three metals W, Pt and Ag after the surfaces have been heat cleaned in a vacuum and then exposed to a number of adsorbing gases. For W, which was measured earlier, the adsorbing gases tested were N_2, CO_2, CO, H_2O, O_2, and D_2.[7] (Only the last two were tested with He.) For Pt, the gases were CO, CO_2 and O_2, and for Ag the gas was O_2.
5. When compared to the heat cleaned surface, the adsorbents increase TMAC about 50%.
6. EAC for Ne is between 2 and 3 times EAC for He on all surfaces, both with and without adsorbants. Lengthy exposure to the atmosphere produces a surface for which this does not apply.
7. Lengthy exposure to the atmosphere produces a surface with accommodation coefficients that are considerably higher than those for a surface briefly exposed to 10 Torr of adsorbing gas. The "atmospheric" surface is stable in the presence of vacuum, limited heating[5] and a glow discharge.

VI. ACKNOWLEDGMENTS

The author gratefully acknowledges the support of the U.S. Office of Naval research and the help of students, Hal Fulton, Frank Lacy, and Tom Harley, in taking and analyzing the data.

Table I. Tangential momentum accommodation coefficients (TMAC) and energy accommodation coefficients (EAC) on polycrystalline Pt and Ag after cleaning by heating in a vacuum and after exposure to various gases. For the surfaces that are grouped together, the + or - values indicate the scatter in the data. This scatter is estimated to be within the absolute error in the individual measured values. If the entry in the table represents a measurement on a single surface, the + or - value is obtained from the estimated accuracy of the velocity and absorption data.

Test Gas	Surface	TMAC	EAC	Sum
Ne	Clean Pt, or N_2 on Pt	0.57 ± 0.06	0.20 ± 0.05	0.78 ± 0.06
He	Clean Pt, or N_2 on Pt	0.55 ± 0.07	0.07 ± 0.03	0.62 ± 0.07
Ne	O_2, CO, or CO_2 on Pt	0.75 ± 0.03	0.47 ± 0.05	1.21 ± 0.03
He	O_2, CO, or CO_2 on Pt	0.76 ± 0.20	0.24 ± 0.08	1.00 ± 0.12
Ne	Pt exposed to atmosphere	0.84 ± 0.08	0.64 ± 0.08	1.48 ± 0.08
He	Pt exposed to atmosphere	0.64 ± 0.08	0.60 ± 0.08	1.24 ± 0.08
Ne	Clean Ag, N_2 or CO on Ag	0.70 ± 0.06	0.39 ± 0.03	1.09 ± 0.07
He	Clean Ag, N_2 or CO on Ag	0.79 ± 0.03	0.13 ± 0.03	0.92 ± 0.04
Ne	O_2 on Ag	0.76 ± 0.08	0.48 ± 0.08	1.24 ± 0.08
He	O_2 on Ag	0.96+.04-0.21	0.16+.08-.04	1.12 ± 0.10
O_2	O_2 on Pt	0.88 ± 0.10	0.84 ± 0.10	1.72 ± 0.10
O_2	O_2 on Ag	0.92 ± 0.10	0.76 ± 0.08	1.68 ± 0.10
CO	CO on Pt	0.92 ± 0.10	0.64 ± 0.08	1.56 ± 0.10
CO	CO on Ag	0.96 ± 0.10	0.76 ± 0.08	1.72 ± 0.10
N_2	N_2 on Pt	0.80 ± 0.08	0.72 ± 0.08	1.52 ± 0.08
N_2	N_2 on Ag	0.80 ± 0.08	0.68 ± 0.08	1.48 ± 0.08

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LEGEND FOR FIGURES

Fig. 1 (A) and (B). Scanning electron microscope photographs of the inner surface of the Pt tube before and after heating; (C) and (D), Photographs of the silver surface before and after heating.

Fig. 2. Reduced sound abosrption and inverse sound velocity in the Pt tube as a function of the reduced parameter r_ω. ★ - heat cleaned surface, average of three runs, ● - surface exposed to 10 Torr of N_2, ☆ - surface exposed to 10 Torr of CO, ▣ - surface exposed to 10 Torr of CO_2, ◎ - surface exposed to 10 Torr of O_2, ◉ - surface exposed to atmosphere. The curves drawn through the points were calculated using the EAC and TMAC values in Table I.

Fig. 3. Reduced sound absorption and inverse sound velocity in the Ag tube as a function of the reduced parameter r_ω. Symbols have the same designation as in Fig. 2. The curves were calculated using EAC and TMAC values from Table I.

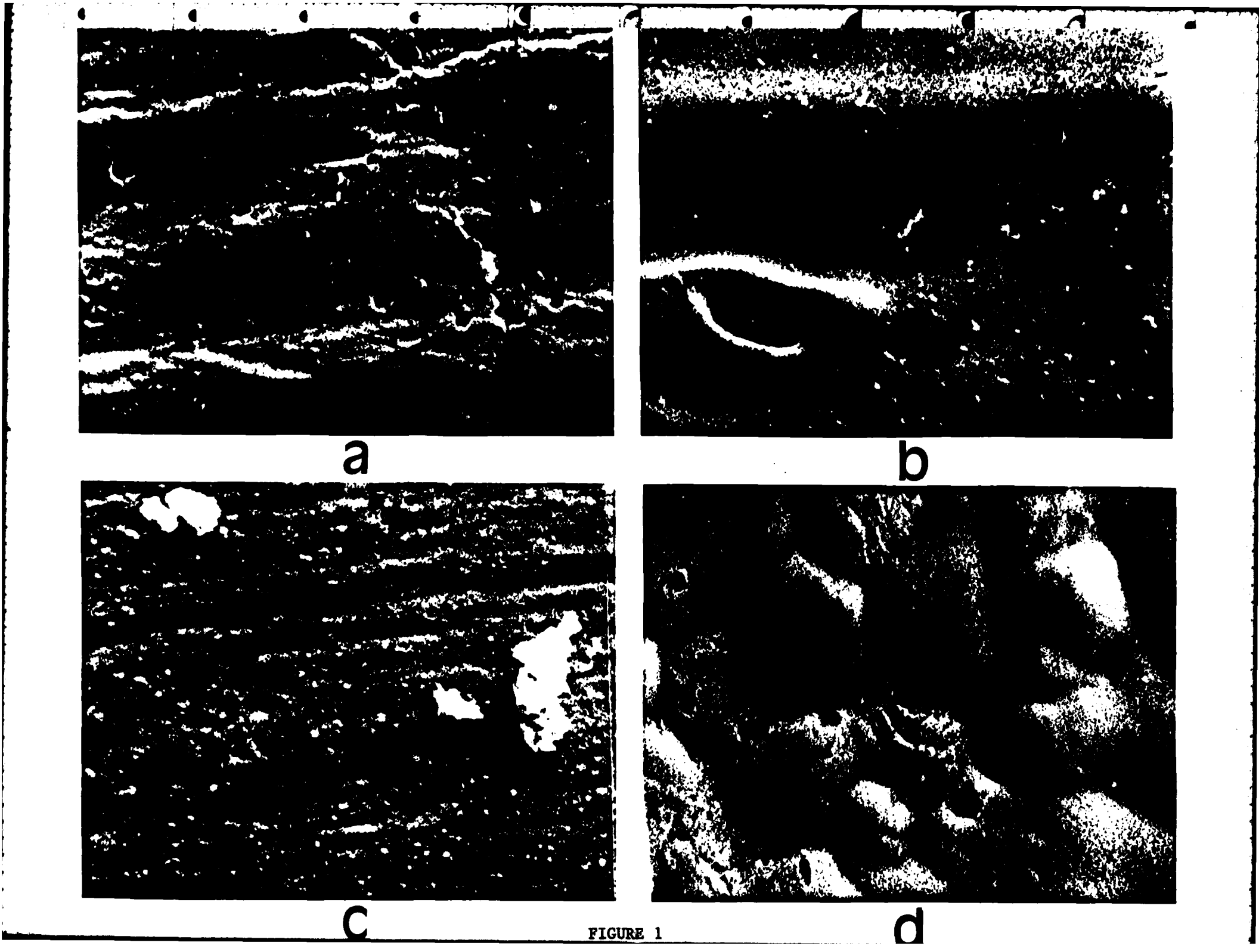
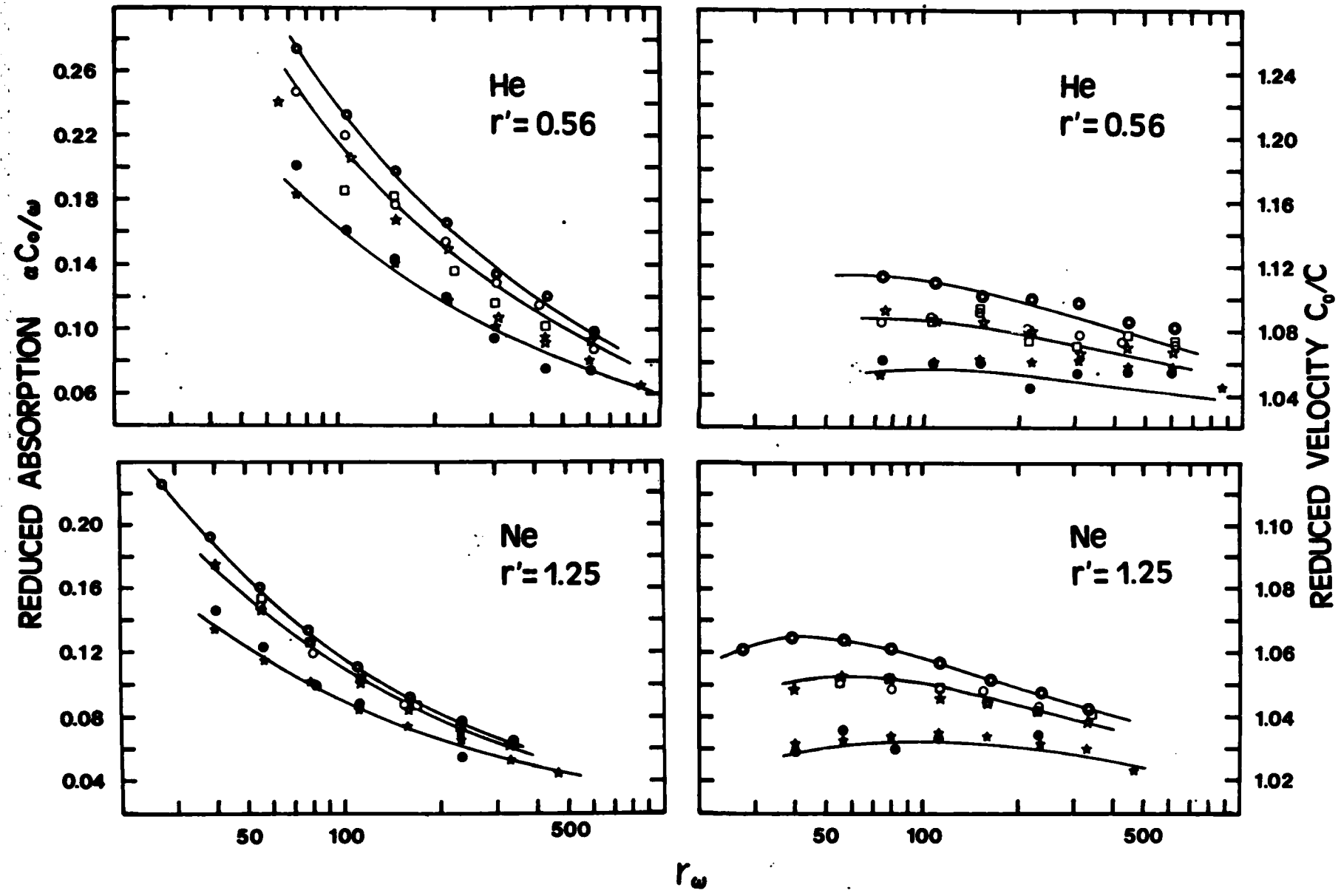


FIGURE 1


He
r'= 0.56
He
r'= 0.56
Ne
r'= 1.25
Ne
r'= 1.25
REDUCED ABSORPTION αC₀/ω
REDUCED VELOCITY C₀/C
0.26
0.22
0.18
0.14
0.10
0.06
1.24
1.20
1.16
1.12
1.08
1.04
0.20
0.16
0.12
0.08
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1.06
1.04
1.02
50
100
500
50
100
500
rω

FIGURE 2

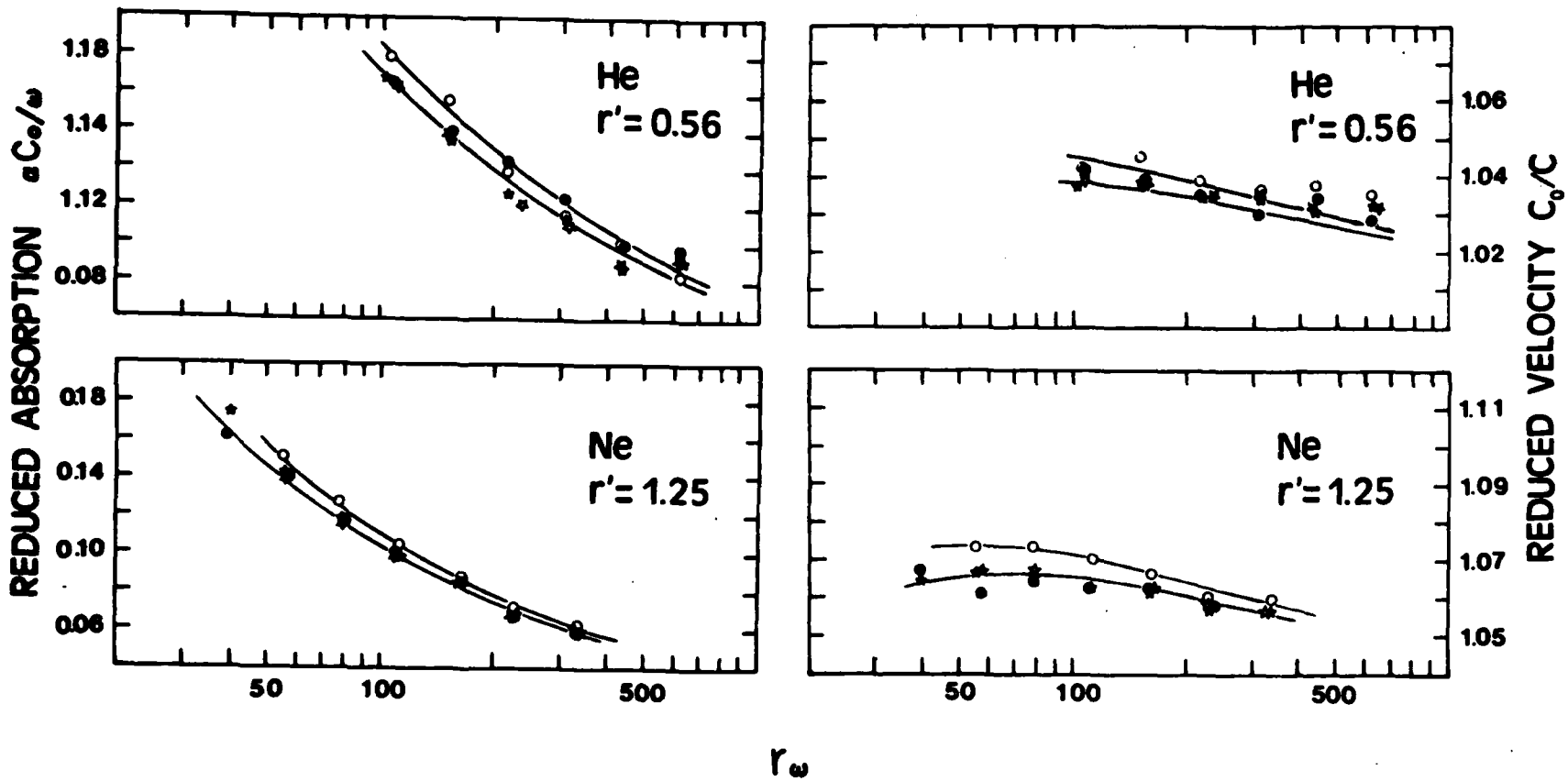
REDUCED ABSORPTION αC₀/ω
1.18
1.14
1.12
0.08
He
r'= 0.56
He
r'= 0.56
1.06
1.04
1.02
0.18
0.14
0.10
0.06
Ne
r'= 1.25
Ne
r'= 1.25
1.11
1.09
1.07
1.05
50
100
500
50
100
500
rω
REDUCED VELOCITY C₀/C

FIGURE 3